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TRIPLY DIFFERENTIAL STUDIES OF ATOMIC AND MOLECULAR  
PHOTOIONIZATION USING..(U) NATIONAL BUREAU OF STANDARDS  
WASHINGTON DC NATIONAL MEASUREME.. A C PARR ET AL.

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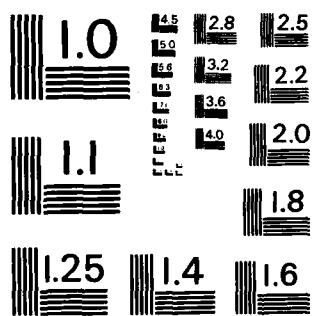
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Basic studies of photoionization processes in atoms and molecules have been carried out using triply differential (differential in incident wavelength, ejected electron energy, and ejection angle) photoelectron spectroscopy with synchrotron radiation as the excitation source. Measurements have been con- ducted in the vacuum ultraviolet wavelength range up to $h\nu \approx 35$ eV on a variety of atomic and molecular systems. Photoelectron branching ratios (partial photoionization cross sections) and photoelectron angular distributions were		

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20. Abstract, Continued.

obtained for all accessible states. Publications on  $C_2N_2$ ,  $SO_2$ ,  $HCN$ ,  $CH_3CN$ , and  $BF_3$  were published or submitted for publication during the last year. The work on  $BF_3$  included extensive comparison with theoretical calculations, which revealed unexpected aspects of the dynamics of shape resonances in molecular photoionization. During the last year, the main effort has been placed on completion of a major new instrument for these studies. A second generation machine, capable of 100-1000 increase in sensitivity and much higher resolution than the initial instrument, has been installed and prepared for operation. This will enable the project to address important measurements which otherwise lay beyond the capabilities of other existing instruments in this field. Opportunities for future directions of this project have been tentatively identified.



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ANNUAL SUMMARY REPORT

TRIPLY DIFFERENTIAL STUDIES OF ATOMIC AND MOLECULAR PHOTOIONIZATION  
USING SYNCHROTRON RADIATION (Contract No. N00014-84-F-0005)

Principal Investigators:

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Submitted to

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Physics Division (Code 412)  
Department of the Navy  
Arlington, VA 22217

Attn: Prof. Douglas Klein

Submitted by

National Measurement Laboratory  
National Bureau of Standards  
Gaithersburg, MD 20899  
3 July 1984

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## I. SCIENTIFIC PROBLEM

The research supported by this contract involves basic studies of vacuum ultraviolet (VUV) photoionization processes in atoms and molecules. Using advanced experimental approaches, described in Section II, we seek to develop new insight and basic data concerning the spectroscopy and dynamics of atomic and molecular photoionization by observing complementary observables, e.g., partial photoionization cross sections, photoelectron branching ratios, and photoelectron angular distributions. These studies have led to novel measurements and to new insight into photoionization mechanisms, particularly regarding the effects of shape resonances and autoionization on vibrational ionization channels.

In addition to our prime goal of developing a clear understanding of photoionization spectroscopy and dynamics, this work impacts on at least three other areas: First, we seek to develop new probes of the photoionization process. In recent annual periods, this project has introduced the techniques of fluorescence polarization spectroscopy of molecules and photoelectron-photoion coincidence spectroscopy of clusters formed in a supersonic expansion. At the present time, attention is focussed on completion of a new, high-resolution angle-resolved, dual photoelectron spectrometer system (discussed below). Future work will be aimed at such experiments as electron-electron coincidence studies, VUV photoionization of laser-excited states, and photoelectron-photoion coincidence studies of molecular fragmentation using synchrotron radiation. Second, this project produces data crucial for testing theoretical predictions and, thus, contributes to the development of realistic theories of atomic and molecular photoionization. Third, the data produced by this project contributes to characterizing the alternative pathways by which radiation interacts with matter, and hence contributes to the macroscopic modeling of such microscopic interactions.

## II. SCIENTIFIC AND TECHNICAL APPROACH

The main experimental approach used in this work involves triply differential photoelectron measurements using synchrotron radiation as the continuously tunable source of ionizing radiation. By triply differential photoelectron studies, we mean that photoelectron intensity measurements are made as a function of three completely independent variables — the incident wavelength,  $\lambda$ , the kinetic energy of the ejected electron,  $T$ , and the angle of ejection,  $\theta$ , relative to the polarization direction of the light. Variation of  $\lambda$  permits the systematic mapping of photoionization properties throughout the vacuum-ultraviolet wavelength range, including probing important spectral features such as autoionizing states, shape resonances, and near-threshold phenomena. Variation of  $T$  permits the selection of particular electronic-vibrational-(rotational) states formed in the photoionization process. Hence, by monitoring the whole manifold of final states (different  $T$ 's), one can determine the effect of alternative photoionization mechanisms (selected by choosing  $\lambda$ , as discussed above) on relative probabilities of forming the various final states. Variation of  $\theta$  permits the characterization of the angular distribution of photoelectrons for each final state and  $\lambda$ . In photoionization of free atoms and molecules the angular dependence of photoelectron intensity has the simple form

$$\frac{d\sigma}{d\theta} = \frac{\sigma_{\text{tot}}}{4\pi} [1 + \beta P_2(\cos \theta)],$$

where  $\sigma_{\text{tot}}$  is the integrated cross section and  $\beta$  is called the asymmetry parameter. Therefore, the angular distribution as a function of  $(T, \lambda)$  can be characterized by  $\beta$ , which in turn can be measured by recording peak strengths at as little as two angles. While it is true that most applications of photoionization data hinge primarily on partial cross sections (or branching ratios), angular distributions play an indispensable role in testing theoretical models and in the fundamental understanding of the photoionization process. Hence, both branching ratios and  $\beta$ 's are critical to sound scientific study of photoionization processes.

In addition, this project introduced two techniques to address new types of problems in molecular photoionization. These are fluorescence polarization spectroscopy of molecules and photoelectron-photoion coincidence spectroscopy on selected clusters formed in mixtures in supersonic expansions. Expertise in these techniques is available for use in future research as the need arises. Further development of our measurement capabilities is under consideration but is also subject to staffing levels. Experimental approaches of interest include electron-electron coincidence measurements of double electron ejection, VUV photoionization of laser excited states, and photoelectron-photoion coincidence studies of molecular fragmentation using synchrotron radiation.

### III. PROGRESS

The scientific accomplishments of this program during the last contract period are reflected, in part, in the papers, abstracts of contributed talks, and invited lectures listed in Section IV. In particular papers 20, and 23-26 were previously submitted, but appeared in this contract period. Papers 27-30 are new submissions in this contract period. In addition, abstracts 31-33 and invited talks 26-32 were added during this contract period.

The highlights of the past year, not all of which are represented by the papers, abstracts, and talks, can be summarized in six categories: First, and most significant for the future of the program, we have devoted the major portion of our research effort to the completion of a new generation double electron spectrometer system for triply differential photoionization studies. This new system has been under development for over two years and will yield two to three orders of magnitude improvement in sensitivity/resolution over our previous instrument and others that have appeared since our initial work in this field. This is crucial for addressing the most significant problems in the field now and in the future. The most important problems frequently require high photon resolution and detector sensitivity to probe typically narrow autoionizing resonances and high electron energy resolution to resolve vibrational modes in molecules more complex than first row diatomics. The steps taken during the last year include: (a) installation of the instrument at the NBS SURF-II synchrotron radiation facility in June, 1983; (b) solution of special magnetic shielding problems caused by the high magnetic field (2 Gauss) environment of the electron storage ring; (c) refinement of the electrical shielding used to protect the collision zone from electric fields caused by the voltages applied to the hemispherical analyzers; (d) completion of the sophisticated software required to interface the instrument, automate the data taking, and analyze the raw data; (e) development of calibration procedures to correct for the analyzers' electron transmission function and a small angular correction



factor; (f) initial data acquisition. Most of this instrument development work is described in paper 28. Application of the instrument to our scientific objectives is beginning at this time. Second, data on  $N_2$ -like molecular environments, taken with our first-generation instrumentation, was analyzed and published or submitted for publication. This included studies on  $C_2N_2$ , HCN, and  $CH_3CN$  described in papers 24, 26, and 27. The objective of this work was to trace the effects of chemical environment on the shape-resonant features known to exist in  $N_2$ . Third, a major study on triply differential photoelectron measurements of valence shell photoionization in  $BF_3$  was prepared for publication. This work, described in paper 29, has been accepted for publication in Phys. Rev. A. It contains a comparison of our data with the only existing theoretical work on the medium-sized molecule  $BF_3$ . This comparison showed that the  $e'$  shape resonance, well known in inner shell spectra, appears in certain valence shell channels but is absent in others. This reinforces the pattern we and others have observed that valence shell photoionization dynamics are more complicated than inner shell dynamics due to continuum-continuum coupling, autoionization, vibrational effects, etc. This study adds an important example to the presently intense study of "missing" shape resonances in valence shell spectra and the search for the responsible dynamical effects. Fourth, we have nearly completed preparation of a study of electric field effects on molecular photoionization, using fluorescence techniques. In a novel study of the effects of electric fields on excited-state thresholds, we have provided evidence for the non-hydrogenic nature of observed threshold shifts caused by short-range forces in the molecular field. Several measurements of this and related types, taken earlier, remain to be analyzed and published. Fifth, a review article (paper 30) associated with invited talk 27 was completed. It gave an overview of shape resonance effects in molecular fields, including several aspects introduced by this project. Sixth an extensive experimental program, stressing the unique high resolution/sensitivity capabilities of our new instrument, has been planned for the coming year. Also, plans for new experimental approaches, mentioned in Sections I and II, have been developed.

#### IV. PUBLICATIONS

The papers, abstracts of conference presentations, and invited talks prepared as part of this ONR program are listed on the following pages. Papers 20, 23-26 were previously submitted, but appeared during his contract period. Papers 27-30 are new submissions in this contract period. In addition, conference abstracts 31-33 and invited talks 26-32 are new in this period.

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24. J. L. Dehmer, A. C. Parr, S. H. Southworth, and D. M. P. Holland, "Triply Differential Photoelectron Studies of Autoionization and Shape-Resonance Effects in Molecular Photoionization," Annual DEAP Meeting, 23-25 May 1983, Boulder, CO, Bull. Am. Phys. Soc. 28, 809 (1983).
25. E. D. Poliakoff, J. L. Dehmer, A. C. Parr, and G. E. Leroi, "Fluorescence Polarization as a Probe of Molecular Autoionization," *ibid.*, p. 809.
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31. A. C. Parr, S. H. Southworth, J. L. Dehmer, and D. M. P. Holland, "Angle Resolved Photoelectron Spectrometers," 3rd National Conference on Synchrotron Radiation Instrumentation, Brookhaven National Laboratory, Upton, Long Island, New York, 12-14 September 1983, Book of Abstracts.
32. J. L. Dehmer, "Shape Resonances in Molecular Fields," National Meeting of the American Chemical Society, St. Louis, Missouri, 8-13 April 1983, Book of Abstracts, p. Phys.-2.
33. S. H. Southworth, A. C. Parr, J. L. Dehmer, and D. M. P. Holland, "Angle-Resolved Photoelectron Study of the Valence Levels of  $\text{BF}_3$ ," XVth Annual Meeting of the DEAP, American Physical Society, Storrs, CT, 30 May - 1 June 1984, Bull. Am. Phys. Soc. 29, 800 (1984).



INVITED TALKS, COLLOQUIA, AND SEMINARS

1. J. L. Dehmer and Dan Dill, "Shape Resonances in Molecular Photoionization," Plenary talk presented at Molecular Spectroscopy and Dynamics with Synchrotron Radiation-A European Workshop, Maria Laach, West Germany, September 29-October 1, 1980.
2. A. C. Parr, "Current Research at NBS Using Synchrotron Radiation at SURF-II," Invited talk presented at the Sixth Conference on the Application of Accelerators in Research and Industry, Denton, TX, November 3-5, 1980.
3. J. L. Dehmer, "Potpourri of Current and Future Studies of Molecular Photoionization-Synchrotron Radiation, Supersonic Jets, and Multiphoton Ionization," Chemistry Department Colloquium, Boston University, Boston, MA, 13 April 1981.
4. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Photoelectron-Photoion Coincidence Spectroscopy of Atomic Clusters and Fluorescence Polarization Analysis," Atomic and Molecular Science Seminar, Argonne National Laboratory, Argonne, Illinois, 6 May 1981.
5. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Molecular Spectroscopy Division Seminar, National Bureau of Standards, Gaithersburg, MD, 21 May 1981.
6. E. D. Poliakoff, "Alignment of Molecular Ions Produced by Photoionization," Seminar on Collision Experiments in Their Theoretical Frame (Fano Workshop), The University of Chicago, Chicago, IL, 23 May 1981.
7. A. C. Parr, "Status of Programs at NBS SURF-II," National Synchrotron Instrumentation Conference, Cornell University, 15-17 July 1981.
8. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Physics Department Colloquium, Georgetown University, Washington, D.C. 6 November 1981.
9. A. C. Parr, "Triply Differential Photoelectron Spectrometry of Atoms and Simple Molecules," Center Colloquium, Center for Absolute Physical Quantities, National Bureau of Standards, January 6, 1982.
10. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron-Photoion Coincidence Studies of Clusters," Brookhaven National Laboratory, December 2, 1981.
11. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron-Photoion Coincidence Studies of Clusters," Exxon Research Laboratory, January, 1982.
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INVITED TALKS, COLLOQUIA, AND SEMINARS, Continued

13. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron-Photoion Coincidence Studies of Clusters," Department of Chemistry, Boston University, January 1982.
14. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron-Photoion Coincidence Studies of Clusters," Department of Chemistry, University of Pennsylvania, February, 1982.
15. A. C. Parr and H. M. Rosenstock, "Resonance, Autoionization, and Kinetic Effects in Photoionization," Invited talk presented at the 30th Annual Conf. on Mass Spectrometry and Allied Topics, Honolulu, June 6-11, 1982.
16. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Department of Physics Colloquium, University of Alabama, March 31, 1982.
17. J. L. Dehmer, "Molecular Photoionization Dynamics - Progress and Prospects," Physics Colloquium, University of Chicago, May 27, 1982.
18. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," California Institute of Technology, June 14, 1982.
19. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," University of California, Santa Barbara, June 15, 1982.
20. J. L. Dehmer, "Overview of Experimental and Theoretical Studies of Resonance Processes in Molecular Photoionization by Single-Photon and Multiphoton Excitation," Gordon Research Conference on Electron Spectroscopy, Wolfeboro, New Hampshire, 19 July, 1982.
21. J. L. Dehmer, D. Dill, and A. C. Parr, "Photoionization Dynamics of Small Molecules," NATO Advanced Study Institute on Photophysics and Photochemistry in the Vacuum Ultraviolet, Lake Geneva, WI, 15-28 August 1982.
22. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Uppsala University, 17 August 1982.
23. J. L. Dehmer, "Resonant Processes in Molecular Photoionization," Meeting of the American Physical Society (Division of Condensed Matter Physics Symposium), Los Angeles, CA, 24 March 1983.
24. A. C. Parr, "Studies of Ion Fragmentation and Molecular Photoionization Using Photoelectron Spectroscopy," JILA Colloquium, Boulder, CO, 8 April 1983.
25. A. C. Parr and R. Stockbauer, "Photoelectron Spectroscopy Studies in Recent Years at NBS," Annual Meeting of the American Society for Mass Spectrometry, Boston, MA, 12 May 1983.

INVITED TALKS, COLLOQUIA, AND SEMINARS, Continued

26. A. C. Parr, "Angle Resolved Photoelectron Spectrometers," 3rd National Conference on Synchrotron Radiation Instrumentation, Brookhaven National Laboratory, Long Island, NY, 12-14 September 1983.
27. J. L. Dehmer, "Shape Resonances in Molecular Fields," Symposium on Resonances in Electron-Molecule Scattering, van der Waals' Complexes, and Reactive Chemical Dynamics, National Meeting of the American Chemical Society, St. Louis, MO, 8-13 April 1983.
28. A. C. Parr, "Studies of Resonances in Molecular Photoionization Using Synchrotron Radiation," Physics Colloquium, University of Alabama, 9 March 1984.
29. A. C. Parr, "Studies of Resonances in Molecular Photoionization Using Synchrotron Radiation," BESSY Synchrotron Radiation Facility, Berlin, W. Germany, 12 April 1984.
30. A. C. Parr, "Studies of Resonances in Molecular Photoionization Using Synchrotron Radiation," LURE Synchrotron Radiation Facility, Orsay, France, 16 April 1984.
31. A. C. Parr, "Overview of Research at NBS Using Synchrotron Radiation at SURF-II," Daresbury Laboratory, England, 14 May 1984.
32. J. L. Dehmer, "Resonant Processes in Molecular Photoionization," Eighth Conference on the Applications of Accelerators in Research and Industry, Denton, TX, 12-14 November 1984.

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